*Indian J. Phys.* 77A (2), 181–186 (2003)

AN INTERNATIONAL JOURNAL IJPA

# Effect of NDOS structure on the transition temperature and resistance anomaly in impure Zr–Cu metallic glasses

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**Abstract** : The suppression of superconducting transition temperature and the anomaly in the resistance peak of $Zr_{50}Cu_{50}$ doped with dilute anti-ferromagnetic impurities have been studied The change in normal density and the resistivities are calculated for 11 ppm of Mn in $Zr_{50}Cu_{50}$. The theoretical results agree with the reported experimental values qualitatively justifying the model proposed.

**Keywords** NDOS structure, transition temperature, Zr–Cu metallic glasses

**PACS Nos.** : 74.62.Bf, 74.70.Ad, 74.25.Fy

## 1. Introduction

With the discovery of high temperature superconductivity, there was a sharp decline in the interest on the superconductivity of metallic alloys for obvious reasons of their low $T_c$ values. But some seminal experimental results on disordered Zr–Cu alloys showing a new resistance anomaly [1,2] near the superconducting transition temperature have revived the interest on the study of these alloys. The anomalies in the resistivity for Zr-Cu metallic glasses containing dilute magnetic impurities consist in the peak in resistance which can reach up to 10 μΩ cm above the usual superconducting fluctuation region. Many such anomalous properties have been accounted for weak localizations, but the question of origin of these effects are still highly controversial and not yet properly understood. We therefore in the present work, reexamine the properties of these alloys theoretically by considering the BCS Hamiltonian along with the *s-d* exchange interaction to investigate how the conduction electrons are affected by the exchange interaction and the cooper pair potential in dilute Zr–Cu alloys. The situation is treated in a way similar to the Kondo effect in dilute magnetic alloys in presence of the BCS interaction.

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## 2. Theory

We expect that in the case of dilute magnetic superconducting alloys, if the *s-d* exchange interaction is anti-ferromagnetic (negative) there appear some correlated state or a kind of bound state at low temperature along with the usual cooper pair states. The problem is solved in a self-consistent way by taking correlation between electrons.

The Hamiltonian for Zr–Cu system which modify the BCS Hamiltonian due to presence of the *s-d* exchange interaction between the conduction electrons and localized moments of the impurity atom [3] is

$$\begin{aligned} H = {} & \sum_{k\sigma} \varepsilon_k c^\dagger_{k\sigma} c_{k\sigma} \\ & + \Delta\left(\sum_l C_{-l\downarrow} C_{l\uparrow} + \sum_{l'} C^\dagger_{l'\uparrow} C^\dagger_{-l'\downarrow}\right) \\ & - \frac{J}{2N} \sum_{ll'} \left\{\left(C^\dagger_{l\uparrow} C_{l'\uparrow} - C^\dagger_{l\downarrow} C_{l'\downarrow}\right) S_z \right. \\ & \left. + C^\dagger_{l\uparrow} C_{l'\downarrow} S_- + C^\dagger_{l\downarrow} C_{l'\uparrow} S_+ \right\}, \end{aligned} \tag{1}$$

where
$$\begin{aligned} \Delta & = -V \sum_k \left\langle C^\dagger_{k\uparrow} C^\dagger_{-k\downarrow} \right\rangle \\ & = -V \sum_k \left\langle C_{k\uparrow} C_{-k\downarrow} \right\rangle . \end{aligned} \tag{2}$$

$C^{\dagger}_{ks}$ and $C_{ks}$ are the creation and annihilation operators of the conduction electrons with wave vector $k$ and spin $s$. $\varepsilon_k$ is the one electron energy, $\Delta$ is the mean field BCS gap defined with cooper-pair potential $V$.

$S_z$ and $S_\pm$ are the components of the spin operator associated with the impurity and $J$ is the strength of the exchange interaction.

## 3. Methodology

To investigate the competing effects of BCS interaction and the $s$-$d$ exchange interaction.

We introduce the Green's functions [4]

$$G_{kk'}(\omega)=\langle\langle C_{k'\uparrow};C^{\dagger}_{k\uparrow}\rangle\rangle, \tag{3}$$

$$\Gamma_{kk'}(\omega)=\Gamma^{(1)}_{kk'}(\omega)+\Gamma^{(2)}_{kk'}(\omega) = \langle\langle C_{k'\uparrow}S_z+C_{k'\downarrow}S_-;C^{\dagger}_{k\uparrow}\rangle\rangle, \tag{4}$$

$$G^{(1)}_{kk'}(\omega)=\langle\langle C^{\dagger}_{-k'\downarrow};C^{\dagger}_{k\uparrow}\rangle\rangle, \tag{5}$$

$$F_{kk'}(\omega)=\langle\langle C^{\dagger}_{-k'\downarrow}S_z+C_{k'\uparrow}S_-;C^{\dagger}_{k\uparrow}\rangle\rangle. \tag{6}$$

Now, we set up the equation of the motion to be satisfied by these functions in the usual way and we have

$$(\omega+\varepsilon_{k'})G^{(1)}_{kk'}(\omega)-\Delta G_{kk'}(\omega) -\frac{J}{2N}\sum_l F_{lk'}(\omega)=0, \tag{7}$$

$$(\omega-\varepsilon_{k'})G_{kk'}(\omega)-\Delta G^{(1)}_{kk'}(\omega)_{kk'} +\frac{J}{2N}\sum_{l'} F_{lk'}(\omega)=\frac{1}{2\pi}\delta_{kk'}, \tag{8}$$

$$(\omega-\varepsilon_{k'})\Gamma_{kk'}(\omega)-\Delta F_{kk'}(\omega) +\frac{J}{N}\left(n_{k'}-\frac{1}{2}\right)\sum_{l'}\Gamma_{kk'}(\omega) +\frac{J}{2N}\left(\frac{3}{4}-m_{k'}\right)\sum_{l'}G_{kl}(\omega)=0, \tag{9}$$

$$(\omega+\varepsilon_{k'})F_{kk'}(\omega)+\Delta\Gamma_{kk'}(\omega) -\frac{J}{N}\left(n_{k'}+\frac{1}{2}\right)\sum_{k'}F_{kk'}(\omega) +\frac{J}{2N}\left(\frac{3}{4}-\frac{m_{k'}}{3}\right)\sum_{k'}G^{1}_{kk'}(\omega)=0, \tag{10}$$

where $n_{k'}=\sum_l\langle C^{\dagger}_{l\uparrow}C_{k'\uparrow}\rangle$,

$m_{k'}=3\sum\langle C^{\dagger}_{l\uparrow}C_{k'\downarrow}S_-\rangle$.

For $S=\frac{1}{2}$, we use the relations

$$S_\pm S_z=\pm\frac{1}{2}S_\pm, S_zS_\pm=\pm\frac{1}{2}S_\pm,$$
$$S_+S_-=\frac{3}{4}+S_z-S_z^2. \tag{11}$$

For solving the above equations from (11), we must approximate higher order Green's functions and combine the operators in its average value. In doing so, such combinations that conserve the total spin do not vanish. For example, $\langle C^{\dagger}_{k\uparrow}C_{k'\uparrow};S_+\rangle$ should vanish but $\langle C^{\dagger}_{k\uparrow}C_{k'\uparrow};S_z\rangle$ should not. Thus, we put

$$\langle C_{k'\uparrow}C^{\dagger}_{l\uparrow}C_{l'\downarrow}S_-;C^{\dagger}_{k\uparrow}\rangle=\langle C_{k'\uparrow}C^{\dagger}_{l\uparrow}\rangle\langle C_{k\downarrow}S_-;C^{\dagger}_{k\uparrow}\rangle +\langle C^{\dagger}_{l\uparrow}C_{l'\downarrow}S_-\rangle\langle C_{k'\uparrow};C^{\dagger}_{k\uparrow}\rangle, \tag{12}$$

$$\langle C_{k'\uparrow}C^{\dagger}_{l\downarrow}C_{l'\uparrow}S_+;C^{\dagger}_{k\uparrow}\rangle=\langle C^{\dagger}_{l\downarrow}C_{l'\uparrow}S_+\rangle\langle C_{k'\uparrow};C^{\dagger}_{k\uparrow}\rangle -\langle C^{\dagger}_{l\downarrow}C_{k'\uparrow}S_+\rangle\langle C_{l'\uparrow};C^{\dagger}_{k\uparrow}\rangle, \tag{13}$$

$$\langle C_{k'\downarrow}C^{\dagger}_{l\downarrow}C_{l'\uparrow}S_z;C^{\dagger}_{k\uparrow}\rangle=\langle C_{k'\downarrow}C^{\dagger}_{l\downarrow}\rangle\langle C_{l'\uparrow}S_z;C^{\dagger}_{k\uparrow}\rangle -\langle C^{\dagger}_{l\downarrow}C_{k'\downarrow}S_z\rangle\langle C_{l'\uparrow};C^{\dagger}_{k\uparrow}\rangle, \tag{14}$$

$$\langle C_{k'\downarrow}C^{\dagger}_{l\downarrow}C_{l'\downarrow}S_-;C^{\dagger}_{k\uparrow}\rangle=\langle C^{\dagger}_{k'\uparrow}C_{l'\downarrow}\rangle\langle C_{k'\downarrow}S_-;C^{\dagger}_{k\uparrow}\rangle +\langle C_{k'\downarrow}C^{\dagger}_{l\downarrow}\rangle\langle C_{l'\downarrow}S_-;C^{\dagger}_{k\uparrow}\rangle, \tag{15}$$

and also use the following relations from above symmetry of the system

$$\langle C^{\dagger}_{l\uparrow}C_{l'\uparrow}\rangle=\langle C^{\dagger}_{l\downarrow}C_{l'\downarrow}\rangle, \tag{16}$$

$$\langle C^{\dagger}_{l\uparrow}C_{l'\downarrow}S_-\rangle=\langle C^{\dagger}_{l'\downarrow}C_{l\uparrow}S_+\rangle =2\langle C^{\dagger}_{l'\uparrow}C_{l'\uparrow}S_z\rangle =-2\langle C^{\dagger}_{l\downarrow}C_{l'\downarrow}S_z\rangle \tag{17}$$

and $\langle S_z\rangle=0$.

Thus solving the eqs. (7–10), we find by linearizing with respect to $\Delta$

$$G_{kk'}(\omega)=\frac{1}{2\pi}\left\{\frac{\delta_{kk'}}{\omega-\varepsilon_k}-\frac{J^2}{4N(\omega-\varepsilon_k)(\omega-\varepsilon_{k'})} \frac{\Gamma(\omega)}{1+JG(\omega)+\frac{1}{4}J^2F(\omega)\Gamma(\omega)}\right\}, \tag{18}$$

$$\Gamma_{kk'}(\omega)=\frac{1}{2\pi}\left\{\frac{J}{2N(\omega-\varepsilon_k)(\omega-\varepsilon_{k'})}\right\}$$

$$\frac{\left(m_{k'}-\frac{3}{4}\right)[1+JG(\omega)]-\left(n_{k'}-\frac{1}{2}\right)J\Gamma(\omega)}{1+JG(\omega)+\frac{1}{4}J^2F(\omega)\Gamma(\omega)}, \quad (19)$$

$$G^{(1)}_{kk'}(\omega)=\frac{\Delta}{2\pi\left(\omega^2-\varepsilon_k^2\right)}$$

$$\frac{1+JG'(\omega)}{1+JG'(\omega)+\frac{1}{4}J^2\Gamma'(\omega)F'(\omega)}, \quad (20)$$

where $F(\omega)=\frac{1}{N}\sum_k\frac{1}{\omega-\varepsilon_k}$,

$$G(\omega)=\frac{1}{N}\sum_k\frac{n_k-\frac{1}{2}}{\omega-\varepsilon_k},$$

$$\Gamma(\omega)=\frac{1}{N}\sum_k\frac{m_k-\frac{3}{4}}{\omega-\varepsilon_k} \quad (21)$$

and $F'(\omega)=\frac{1}{N}\sum_k\frac{1}{\omega+\varepsilon_k}$,

$$G'(\omega)=\frac{1}{N}\sum_k\frac{n_k-\frac{1}{2}}{\omega+\varepsilon_k},$$

$$\Gamma'(\omega)=\frac{1}{N}\sum_k\frac{\frac{m_k}{3}-\frac{3}{4}}{\omega+\varepsilon_k} \quad (22)$$

**4. Solutions at high and low temperatures**

The high temperature, limit of $m_{k'}$ and $n_{k'}$ may be replaced by their zeroth order quantities because they appear only in higher order perturbation terms. Therefore, $m_{k'}=0$ and $n_{k'}=f_{k'}$ (Fermi function).

Then eq. (18) becomes

$$G_{kk'}=\frac{1}{2\pi}\left[\frac{\delta_{kk'}}{\omega-\varepsilon_k}+\frac{3J^2}{16N}\right.$$

$$\left.\frac{F(\omega)}{(\omega-\varepsilon_k)(\omega-\varepsilon_{k'})(1+JG^{(0)}(\omega))}\right] \quad (23)$$

In particular,

$$\frac{1}{2\pi}\left[G_{kk'}(\omega)\right]^{-1}=(\omega-\varepsilon_k)-\frac{3J^2}{16N}\frac{F(\omega)}{1+JG^{(0)}(\omega)}, \quad (24)$$

where $G^{(0)}(\omega)=\frac{1}{N}\sum_k\frac{f_{k}-\frac{1}{2}}{\omega-\varepsilon_k}=K(\omega)-iL(\omega)$.

$K(w)$ and $L(w)$ are real functions of '$w$' defined by

$$K(\omega)=\frac{1}{2}\sum_k P\frac{f_k-\frac{1}{2}}{\omega-\varepsilon_k}. \quad (25)$$

$$L(\omega)=\pi\rho\left[f(\omega)-\frac{1}{2}\right]. \quad (26)$$

The function $F(w)$ defined by eq. (21) is evaluated and is replaced by pure imaginary constant given by

$$F(\omega)=-\frac{i\pi\rho_0}{N} \quad (27)$$

Here, $\rho_0$ defines the density of the conduction electrons near the Fermi surface. In calculating $K(w)$, we replace the summation over '$k$' by the integration over $\varepsilon_k$ and assume the density of states to be independent of $w$, and cut off the integration at some energy which should be of the order of band width.

Hence,

$$K(\omega)=\frac{\rho_0}{N}\int_{-\infty}^{+\infty}P\frac{f(\varepsilon_k)-\frac{1}{2}}{\omega-\varepsilon_k}d\varepsilon_k$$

$$=\frac{\rho_0}{N}\int_{-D}^{+D}P\frac{f(\varepsilon_k)-\frac{1}{2}}{\omega-\varepsilon_k}d\varepsilon_k$$

$$=-\frac{\rho_0}{N}\int_{-D}^{+D}\frac{1}{\omega-\varepsilon_k}\tanh\left(\frac{\varepsilon_k}{2T}\right)d\varepsilon_k. \quad (28)$$

At $T=0$,

$$K(\omega)=-\left(\frac{\rho_0}{N}\right)\ln(\omega/D). \quad (29)$$

By using the values of $F(w)$ and $G^{(0)}(w)$, we can write eq. (24) as

$$\frac{1}{2\pi}\left[G_{kk'}(\omega)\right]^{-1}=(\omega-\varepsilon_k)$$

$$-\frac{3J^2\rho}{16N^2}\frac{JL(\omega)}{\left[1+JK(\omega)\right]^2+\left[JL(\omega)\right]^2}$$

$$+i\frac{3\pi J^2\rho}{16N^2}\frac{1+JK(\omega)}{\left[1+JK(\omega)\right]^2+\left[JL(\omega)\right]^2}. \quad (30)$$

If $J<0$, and temperature is low enough, the imaginary part of the inverse of the life time of conduction electrons, becomes negative near $w=0$.

This means that the conduction electron states become unstable near the Fermi surface and this instability arises below a critical temperature $T_c$ called Kondo temperature obtained from

$$-\frac{|J|}{2N}\sum_i \frac{n_k - \frac{1}{2}}{\varepsilon_k} = 0. \tag{31}$$

Now, if we consider the case for which $J < 0$ and $T < T_c$, $m_{k'}$ is expected to be large near $\varepsilon_k = 0$.

Let us assume the relation [4]

$$m'_k - \frac{3}{4} = \alpha\left(\frac{n_{k'} - \frac{1}{2}}{\varepsilon_k}\right) \quad \alpha \geq 0. \tag{32}$$

Using (31) and (32), the normal and anomalous Green's functions at low temperature become

$$G_k(\omega) = \sum_{k'} G_{kk'}(\omega) = \frac{1}{2\pi}\frac{\omega}{(\omega - \varepsilon_k)(\omega + i\Delta_1)} \tag{33}$$

and $$G_k^{(1)}(\omega) = \sum_{k'} G_{kk'}^{(1)}(\omega) \quad \frac{\Delta}{2\pi}\frac{\omega}{(\omega^2 - \varepsilon_k^2)(\omega + i\Delta_1)}, \tag{34}$$

where $\Delta_1 = \frac{\pi}{4N}|J|\rho\alpha$.

## 5. Evaluation of NDOS and superconducting $T_c$ and the resistivity near $T_c$ change

The NDOS (normal density of states) near the Fermi surface and the resistivity near $T_c$ are obtained from the Green's functions (30) and (33) and the transition temperature and the drop of transition temperature are obtained from eq. (34) and the NDOS calculations.

The NDOS is defined by

$$\rho(\omega) = -2\sum \mathrm{Im}\langle G_k(\omega)\rangle. \tag{35}$$

The effect on NDOS due to alloying and to suit our interest, we consider the change in NDOS from its unperturbed value,

$$\delta\rho(\omega) = -2\sum \mathrm{Im}\{\langle G_k(\omega)\rangle - G_k^{0N}(\omega)\}, \tag{36}$$

where $G_k^{0N}(\omega) = \mathrm{Lt}_{\Delta\to 0} G_k^0(\omega)$,

$$\delta\rho(\omega) = -\frac{1}{\pi}\sum_k \mathrm{Im}\frac{\Sigma(\omega)}{(\omega - \varepsilon_k)^2},$$

or $$\delta\rho(\omega) = \frac{\rho_0(\varepsilon)}{\pi}\mathrm{Im}\sum(\omega)\frac{\partial}{\partial\omega}F(\omega), \tag{37}$$

where $S(w)$ is the self energy of the system.

At this stage, one needs to make a choice of $\rho_0(\varepsilon)$ under the simplest assumption of square $\rho_0(\varepsilon)$ which becomes independent of $\varepsilon$, $F(w)$ is constant and there is no change in NDOS. Therefore, more general approximation could be to assume a Lorentzian given by [5]

$$\rho_0(\varepsilon) = \rho_0(0)\frac{D^2}{D^2 + \varepsilon^2}, \tag{38}$$

$$F(\omega) = \pi\rho_0(0)\frac{D}{\omega + iD}. \tag{39}$$

The change in NDOS is thus calculated from the self energy obtained from eq. (30) by substituting $L(w) = 0$ and for finite concentration $C = \frac{N_i}{N}$ of the anti-ferromagnetic impurity ($J \leq 0$).

$$\delta\rho(0) = \rho(0) - \rho_0(0)$$

$$= \rho_0(0)\left\{1 \pm \frac{3\pi JC}{16ND\ln(T/T_c)}\right\}.$$

–ve sign is for $T > T_c$ and $J < 0$, +ve sign $T < T_c$ and $J > 0$ (Kondo anomaly) below and above Kondo temperature $T_c$, the transition temperature is obtained from the gap eq. (34)

$$\Delta = -V\sum_k \langle C_{k\uparrow}^{\dagger} C_{-k\downarrow}^{\dagger}\rangle,$$

where the gap correlation function is

$$\langle C_{k\uparrow}^{\dagger} C_{-k\downarrow}^{\dagger}\rangle = i\int_{-\infty}^{+\infty} d\omega \frac{[G^{(1)}(\omega + i\varepsilon) - G^{(1)}(\omega - i\varepsilon)]}{e^{\beta\omega} + 1}$$

Using eq. (29), we obtained

$$\Delta = \frac{\Delta V}{2}\sum_k \frac{\varepsilon_k}{\varepsilon_k^2 + \Delta^2}\tanh\left(\frac{\beta\varepsilon_k}{2}\right)d\varepsilon_k,$$

or $$\Delta = \Delta_0 e^{-\frac{\pi^2}{6}\left(\frac{T}{\Delta_0}\right)},$$

where $\Delta_0 = De^{-\frac{1}{V\rho_0(0)}}$,

hence $T'_c = 1.14\Delta_0 = 1.14De^{-\frac{1}{V\rho_0(0)}}$.

The experimental results suggest that the resistivity shows an anomalous peak near $T'_c$ but above $T'_c$ over a narrow range of 20 $mK$ – 60 $mK$, called the superconducting fluctuation. We evaluate the resistivity for the alloy from the normal state Green's function equation (30).

The static conductivity $s$ is calculated by the formula

$$\sigma = -\frac{2e^2}{3}\int \tau_k v_k \rho \frac{\partial f}{\partial\varepsilon_k} d\varepsilon_k, \tag{40}$$

where $v_k$ is the velocity of the conduction electron with wave vector $k$ and $\tau_k$ is the mean free time.

From eq. (27),

$$\frac{1}{\tau_k} = \frac{3\pi J^2 \rho c}{16N} \frac{1}{1-|J|K(\varepsilon_k)} \quad \text{for } J < 0, \tag{41}$$

$$= \frac{2e^2 \rho v_F^2}{3} \frac{16N}{3\pi J^2 \rho c}$$

$$\int_{-\infty}^{+\infty} \left(1-|J|K(\varepsilon_k)\left(-\frac{\partial f}{\partial \varepsilon_k}\right)\right) d\varepsilon_k$$

$$= \frac{ne^2}{m^*} \frac{16}{3\pi|J|c\rho} \ln(T/T_c'), \tag{42}$$

where $T'_c$ is the alloy transition temperature after impurity doping and $n$ is the total number of conduction electrons.

Thus, $\rho = \frac{3n}{2m^* v_F^2}$, where $m^*$ = density of effective mass of the conduction electrons.

Thus the resistivity,

$$R = \frac{1}{\sigma} = \frac{m^* 3\pi|J|c}{16ne^2} \left[\ln|T/T_c'|\right]^{-1} \tag{43}$$

The new transition temperature $T'_c$ after doping the magnetic impurity is related to $T_c$ of the alloys before doping as

$$\frac{T_c'}{T_c} = \exp\left[-\left(\frac{1}{V\rho(0)} - \frac{1}{V\rho_0(0)}\right)\right], \tag{44}$$

where $\rho_{(0)}$ is the density of states of the conduction electrons near the Fermi surface after impurity doping.

So the resistivity $R$ becomes

$$R = \frac{m^* 3\pi|J|c}{16ne^2} \left[\frac{1}{V\rho(0)} - \frac{1}{V\rho_0(0)} + \ln(T/T_c)\right]^{-1}. \tag{45}$$

## 6. Results and discussion

The suppression of the superconducting transition temperature due to impurity doping is obtained for 11 ppm of Mn in $Zr_{50}Cu_{50}$ from eq. (39) and eq. (43) for $D$ = 0.1 eV, $J$ = 0.62 in the limit $\Delta T_c$ = 40 mK and $V\rho_0(0)$ = 0.568, $T_c$ = 0.92 [6] as was obtained experimentally. The corresponding resistivity ratios $R/R_{1.1K}$ are obtained around the fluctuation region of 20 mK – 60 mK for 11 ppm of Mn in $Zr_{50}Cu_{50}$. The variation $\rho(0)/\rho_0(0)$ with $T \geq 0.47\ T_c$ and the resistivity $R/R_{1.1K}$ are shown in Figures 1 and 2. The resistivity shows a peak maximum which follows $T_c$ over a range of 20 mK – 60 mK above the resistive midpoint of the superconducting transition temperature. This is in good qualitative agreement with the experimental result and

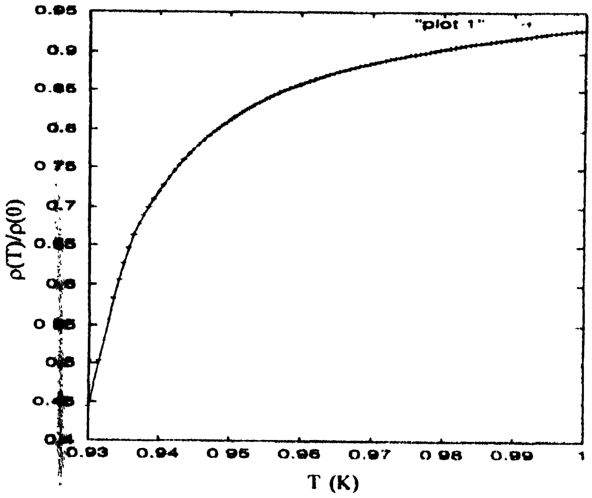


**Figure 1.** Variation of density of states of impure $Zr_{50}Cu_{50}$ superconductor at $T > T_c$ (11 ppm of Mn).

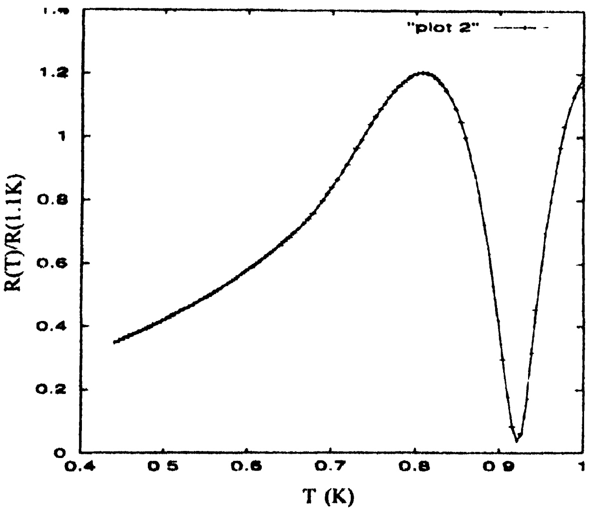


**Figure 2.** Variation of resistivity of impure $Zr_{50}Cu_{50}$ with temperature $T > T_c$.

strongly suggests that the peak arises from the interaction between superconducting fluctuations and dilute magnetic impurities. The experiments suggest that the peak can be quenched by applying moderate magnetic field and shows a strong negative magneto-resistance giving further evidence for the role of interactions between spins and conduction electrons.

## Acknowledgment

One of the authors (A K Mondal) is thankful to the University Grants Commission, Government of India for financial support.

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